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DETERMINATION OF DIFFUSION COEFFICIENTS ON BASIS  
OF ABSORPTION OF BETA-EMISSION

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[Numbers in brackets refer to appended bibliography.]

Knowledge of diffusion characteristics in solid solutions is essential for the solution of a number of problems of preparation of a material with specified properties. The use of artificially radioactive materials facilitated the tracing of these characteristics. During recent years, a great number of works devoted to various methods of measuring diffusion coefficients have appeared which are based on the application of radioactive materials.[1]

However, all known methods have substantial deficiencies. Many methods involve the cumbersome process of cutting the specimen into layers. The use of another group of methods (method of longitudinal cut) is complicated by excessive requirements of precision of distance measuring.

Of interest are absorption methods using the variation of absorption of emission in diffusion. A basic deficiency of various modifications of the absorptive method consists in inaccurate definition of the absorption dependence on distance. Actually Bear's law is accepted for beta-rays, which is completely incorrect in the case of a mixed spectrum and even inaccurate in the case of a simple one. This is the deficiency of the method recently described by P. L. Gruzin and D. F. Litvin [2], containing simultaneously a useful indication as to the expediency of employing gamma-radiation as evidence. One of us developed with the assistance of S. N. Kryukov the method of the "thin layer," which does not require at all the knowledge of absorption laws.[3] However, this method requires the preparation of thin layers, which involves difficulties in the case of fragile materials, and it may in some cases invoke doubts as to whether the structure of the material of the specimen has been retained.

It is of interest to study the possibility of developing an absorption method possessing the advantages of "thin layers" but realized on a "thick layer." The presentation of such a method is the subject of this article. The method should be based on the measurement of the drop in activity produced by the diffusion of the radioactive material within the thick layer. Simultaneously with this value, a certain characteristic of adsorption and radiation diffusion in the material should be used. The adsorption coefficient  $\mu$  of the Bear law does not correctly reflect this phenomenon. The direct measurement of the function  $\phi(x)$  representing the decrease of radiation produced by the penetration of the emitter to a depth  $x$  is rather difficult. The most efficient would be the following integral characteristic of absorption and scattering:

$$A_0 = \int_0^{\infty} \phi(x) dx$$

This quantity is not bound to some assumption on the nature of absorption or scattering. This is easy to determine experimentally inasmuch as it is connected with the emission of the specimen with uniformly distributed radioactive material. The quantity  $A_0/\sqrt{Dt}$  (here  $D$  is the diffusion coefficient and  $t$  is the annealing time) is the expedient dimensionless independent variable on which drop in activity ( $I_t/I_0$ ) should depend.

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If on the surface of a semifinite layer a quantity  $Q$  of matter is deposited, it is known that the concentration  $C$  as a function of time and depth will be expressed in the following way [4]:

$$C(x,t) = \frac{Q}{\sqrt{\pi D t}} e^{-x^2 / 4Dt} \quad (1)$$

In accordance with the above,

$$\frac{I}{I_0} = \frac{1}{\int_0^\infty \frac{1}{\sqrt{\pi D t}} e^{-x^2 / 4Dt} \varphi(x) dx} \quad (2)$$

where  $\varphi = \varphi(0)$  accounts for the reflection effect.

We shall look for a solution for large values of the time. Expanding the exponential function into series, we obtain:

$$\frac{I}{I_0} = \frac{A_0}{\int_0^\infty \sqrt{\pi D t}} + \frac{A_2}{4V\pi(\sqrt{Dt})^3} + \frac{A_4}{32V\pi(\sqrt{Dt})^5} + \dots \quad (3)$$

Here  $A_n$  is the corresponding moment of the functions  $\varphi$ ,  $A_n = \int_0^\infty x^n \varphi(x) dx$ .

For sufficiently large values of the time, the first term of this expansion is satisfactory. In this case,  $D$  may be determined from the graph of  $I/I_0$  versus  $1/\sqrt{t}$ .

The realization of linearity on the graph confirms the realization of the corresponding asymptotic dependence, i.e., the sufficiency of the annealing time assigned. The instant of the occurring linearity depends, of course, on the quantities  $D$  and  $A_0$ .

The quantity  $A_0$  depends on the intensity of absorption and will decrease with it. For this reason, the asymptotic dependence will occur much earlier, in the case of a strongly absorbing emission. Insofar as no assumptions were made as to the nature of  $\varphi(x)$ , in order to shorten the duration of the experiment, we may separate for observation the soft part of the beta-spectrum. This is accomplished by the use of suitable filters.

Finally, we should note that the activation energy may be determined by the following simple method, which also does not require any assumption as to the nature of the absorption. From the concept of dimension it follows that  $I/I_0$  should depend on the product  $Dt$ . Therefore, if at two different temperatures and during different time periods  $I/I_0$  reaches the same value, this means the stability of the product  $Dt$ . From the equality  $D_1 t_1 = D_2 t_2$  it is easy to determine the energy of activation.

We tested the new method of measuring diffusion characteristics in an example of self-diffusion of silver. For this case comparison data are available [3, 5]; besides  $Ag^{110}$  has a complex spectrum which makes the usual version of the absorption method not applicable.

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The silver used was purified by familiar chemical methods.[6] Spectral analysis showed only traces of Cu and Si. Radioactive silver was deposited by electrolysis. The counting of activity was carried out on an end counter. The dimensions of the specimens were  $0.5 \times 0.5 \times 0.1$  cm. The experiments were carried out at five temperatures: 925, 900, 850, 800 and 750°. Diffusive annealing was carried out in a vacuum of the order  $10^{-3}$  mm. Hg. The temperature was kept constant to  $1^\circ$ .

Figure 1 [appended] represents results of direct measurement of decrease of activity with diffusive annealing for two temperatures. To eliminate errors connected with evaporation losses, etc., we recorded the ratio of beta radiation to gamma radiation. To this end the radiation was measured without filter ( $I_\beta + I_\gamma$ ) and with filter ( $I_\gamma$ ) and the ratio  $I = (I_\beta + I_\gamma) - I_\gamma$  /  $I_\gamma = I_\beta / I_\gamma$  was computed. The ordinate axis shows in figure 1 the ratio of the value  $I$  to its value ( $I/I_0$ ) at the initial instant. By varying the thickness of the filter, one can choose a different part of the beta-spectrum. The presented graphs are obtained with an aluminum filter 22 mg/cm<sup>2</sup> thick.

Figure 2 represents in coordinates  $I/I_0$  versus  $1/\sqrt{t}$ , the results of measurement at two temperatures.

For the computation of the diffusion coefficient, the measurement of the quantity  $A_0$  was carried out. To this end the specimen was, after diffusion experiments, fused and flattened in a pressform.  $A_0$  was computed from the formula:

$$A_0 = \Delta \frac{I_\infty}{I_0} \varphi \quad (4)$$

where  $\Delta$  is the thickness of the specimen, and  $I_\infty$  the ratio of its beta- and gamma-activities (the measurements are carried out with the same filter). (The value  $I_0$  was indicated above.)

If we designate by  $m$  the angular coefficient of the asymptotic line  $I/I_0 - 1/\sqrt{t}$ , then it follows from equation (3) and (4) that

$$D = \frac{I_\infty^2 \Delta^2}{I_0^2 m^2}$$

The values of  $D$  computed from this equation are compiled in the following table.

t°C	925	900	850	800	750
D cm <sup>2</sup> / sec	$3.3 \cdot 10^{-9}$	$2.2 \cdot 10^{-9}$	$9.2 \cdot 10^{-10}$	$3.7 \cdot 10^{-10}$	$1.3 \cdot 10^{-10}$

The value of  $D$  corresponding to the obtained results is  $D_0 = 0.53$  cm<sup>2</sup>/sec. and  $Q = 45,000$  cal/mol, which concurs well with data by Johnson [5] ( $D_0 = 0.895$  cm<sup>2</sup>/sec.,  $Q = 45,950$  cal/mol) and by Kryukov and Zhukhovitskiy ( $D_0 = 1.28$  cm<sup>2</sup>/sec.,  $Q = 47,400$  cal/mol).

A direct determination of activation energy from figure 1, based on the constancy of the product  $Dt$  at constant  $I/I_0$ , leads to a value of  $Q = 45,900$  cal/mol).

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The accuracy of the method presented here, as well as of other versions of the absorptive method, is basically determined by the accuracy of the temperature stability during the diffusive annealing. An important peculiarity of this method is the right choice of the filter allowing one to obtain in a short time the asymptotic expression for the drop in activity.

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[Appended figures follow:]

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Figure 1.

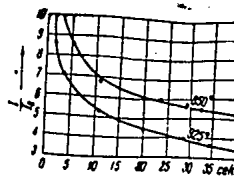
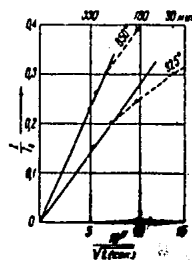


Figure 2.



- E N D -

- 5 -